**New Perspectives on Vacuum High Voltage Insulation. III. Gas Desorption in High Electric Fields**

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To be submitted to – any suggestions?

DRAFT DOCUMENT

February 23, 2012

# ABSTRACT

An examination has been made of gas desorption from unbaked electrodes of copper, niobium, stainless steel and titanium subjected to high voltage in vacuum. It has been shown that large quantities of gas were desorbed from the anode surface during initial high voltage conditioning of the electrodes. The gas was composed of water vapour, hydrogen, carbon monoxide and carbon dioxide plus a high yield of light hydrocarbons. The quantity of gas released was equivalent to many monolayers and consisted mostly of neutral molecules with an ionic component of a few percent.

A number of other experiments completed during that research program examined thermal desorption from the same electrodes for comparison to the desorption that occurred during high-voltage conditioning. Thermal desorption cycles to about 600oC were done with a high electric field present and compared to the same thermal cycle without an electric field. Thermal desorption from an anode with a nearby (3 to 6 mm) cathode at 40 to 50 kV showed much higher desorption of light hydrocarbons compared to desorption without the electric field present. At the same time, hydrogen desorption was reduced by a large amount (up to a factor of ten). It appears that the hydrogen may have combined with surface carbon contamination under the influence of the electric field to produce the hydrocarbons. Residual gas analysis of the gas desorbed during high-voltage conditioning showed similar spectra to those produced when the same electrode was heated with a high electric field present. These observations suggest that there may be complex chemistry at the electrode surface produced by the high electric field. Analysis of desorption products produced during the high-voltage conditioning of the electrostatic deflector of the Tandem Accelerator SuperConducting Cyclotron (TASCC) at Chalk River Laboratories also showed significant quantities of both atomic and molecular hydrogen were desorbed along with water vapour and carbon monoxide, carbon dioxide and many light hydrocarbons.

The thermal desorption tests with a high electric field present also showed that positive ions (and x-rays from secondary electrons from a nearby cathode) were produced during the thermal desorption cycle. The ions and x-rays were correlated with the onset of and an increase in desorption of hydrogen from the metal and were still present in subsequent thermal cycles made without venting the vacuum system. Hydrogen desorption is part of high-voltage conditioning during any of the measurements made that recorded spectral information with a residual gas analyzer. All of these tests were done with no measurable field emission.

These observations support a possible explanation of high-voltage conditioning that is different than previous descriptions. Hydrogen is desorbed from (or near) the surface of the anode under the influence of an electric field and a significant fraction (possibly the atomic fraction) of it is charged. These positive ions are accelerated to the cathode, releasing both secondary electrons (the coefficient of secondary electron emission is above unity for 10’s to many 100’s of keV) and secondary ions and molecules. The electrons are accelerated back to the anode producing x-rays. In parallel with this there appears to be an electrochemical reaction at the anode surface in which the desorbing hydrogen combines with the carbon contamination to produce light hydrocarbons. This appears to be the source of gas that leads to microdischarges that are observed during the initial conditioning of electrodes at high voltage in a vacuum. The desorbed vapour can lead to significant current and if not controlled can lead to a spark. The experiments reported here provide an additional perspective to the very complex processes that occur during dc high-voltage conditioning and suggest some new approaches to electrode material selection and processing of high-voltage equipment.

# 1. INTRODUCTION

High-voltage conditioning of electrodes in a vacuum has remained more an art that a technology since some of the earliest vacuum-high-voltage devices were produced. Various groups use very different approaches to condition high-voltage devices such as spot-knocking, controlled discharges and long periods of conditioning a high-voltage gap as the voltage is increased. It is generally accepted that the major source of high-voltage conditioning is field emission [1,2] from the cathode that eventually leads to either explosive emission of the emitting point as the field-emitted current becomes high or thermal instability of the anode at the location where the electron beam interacts [2]. However, research on materials and surface preparation, especially the use of clean rooms for all operations with the high-voltage electrodes and the vacuum chamber and surface cleaning using pressurized water rinses [3] or a dry ice jet [4] after such treatments as electro- or chemical-polishing of all surfaces have made large improvements in the quality of electrode surfaces. This work has been driven by the laboratories developing superconducting rf cavities and has led to major improvements in the accelerating gradients of rf cavities [5, 6]. In spite of this progress, recent efforts at several laboratories to produce high dc voltages for electron guns to be used with free-electron lasers [7, 8 and 9] have had challenges reaching fairly modest gradients. The problems may not lie with field emission from the cathode as much as generally accepted.

Electrical conductivity and eventual breakdown occurs between two metal electrodes in a vacuum as voltage is increased between them. At least three processes[9,10] occur that contribute to the conduction: field emission of electrons; transfer of microparticles between electrodes; and microdischarges. In parallel with each of these processes there may be an increase in the pressure in the supporting vacuum. Microdischarge activity and the associated pressure increase is often the limiting factor in high-voltage devices such as accelerator tubes and electrostatic analyzers[11]. Microdischarges are self-limiting discharges that occur between unconditioned electrodes as the voltage is increased. They can be observed on an insulated anode or cathode as electrical pulses of many microamperes amplitude and hundreds of microseconds width, i.e., about 3 x 109 electrons or ions per microdischarge. The energy dissipated in a microdischarge is much less than the total energy stored by the capacitance of the electrodes. They have often been ascribed to contaminated electrodes [11] or to a regenerative buildup of positive and negative ions. Chatterton[12] (in 1970) provided a good description of the phenomena and gave a possible explanation. The basic argument was that an ionization process is triggered randomly in the gap producing an electron-ion pair, followed by a release of secondary negative ions at the cathode and secondary positive ions at the anode. If "A" negative ions are produced per positive ion impacting the cathode and "B" positive ions per negative ion impacting the anode, then if AxB is greater than unity, the current will grow regeneratively. However, there must be a continual source of new ions at both surfaces. Experiments[13] have been performed to measure the secondary ion coefficients to demonstrate this as a possible mechanism for contaminated surfaces but not clean surfaces under ultrahigh vacuum conditions. However, gas desorption appears to be the limiting conditioning mechanism in ultra- and extremely-high vacuum systems used for the free-electron laser electron guns [7, 8 and 9].

Both this paper and two previous previous papers [14, 15] report results from an extensive research program on vacuum high-voltage insulation. The first paper [14] provided strong evidence of physical processes occurring at both electrodes that lead to field emission, a key precursor to electrical breakdown. The second paper focused on gas desorption. Evidence was presented [15] that suggested that microdischarges were caused by micro bursts of gas of sufficient density to support a local avalanche discharge. The key experiments that support this thesis were described in reference 15 and are reviewed in this paper because of the importance of these observations in understanding high-voltage conditioning. A number of experiments that measured the properties of thermal desorption of the same electrodes used for the high-voltage experiments were conducted and are described in this paper and several new observations are reported. The experimental work has been described in sufficient detail that it can be easily duplicated. The author has presented his interpretation of this data, aware that there may be further explanations of the complex behaviour.

# 2. EXPERIMENTAL DETAILS

Most of the experiments reported in this paper were done on the test stand shown schematically in Figure 1. The vacuum chamber was made from thick (12-mm) stainless steel to absorb most x-rays and was pumped through a known conductance of 70 l/s as shown. The pressure was measured with ion gauge # 1 on the test stand and ion gauge # 2 after the known conductance. The vacuum pump was a 200 l/s Varian turbo molecular pump backed by a rotary pump. This configuration allowed the measurement of the quantity of gas desorbed during experiments. A standard conflat window enabled measurement of the x-ray and visual activity from the electrodes. The insulators were developed for the electrostatic deflector of the Tandem Accelerator SuperConducting Cyclotron (TASCC) and are described in other publications [16, 17]. Two variable high-voltage power supplies were used, one with a maximum rating of -100 kV, and the other with a maximum rating of ±150 kV. The cable insulation and the ceramic insulator limited the maximum useable voltage to about 110 kV. A residual gas analyzer was used for most of the measurements reported in this paper.

X-ray activity was measured during many experiments with several radiation-survey instruments, scintillation detectors and a high-quality silicon detector. The scintillation detectors provided sensitive temporal response to x-rays while the Geiger counter provided high x-ray sensitivity to field emission. It could measure an increase in x-rays above background that corresponded to measured electron currents of about 50 pA at 50 kV and less than 20 pA at 100 kV.



Figure 1. The basic test stand used to measure gas desorption from electrodes at high voltage.

Figures 2 and 3 illustrate several configurations of a grounded electrode used in place of one of the electrodes shown in Figure 1 for most of the thermal desorption experiments. The electrode was attached to a low-mass stainless steel mount, welded to a light stainless steel tube. A cartridge heater was installed on the atmospheric side of the mount and a thermocouple was inserted into the mount as shown in Figure 2a. The electrode, typically 25 mm diameter by 12 mm thick with radiused corners, was attached to the mount with a stainless steel threaded rod with one side flattened to provide some pumping, and a slot cut across the mount, also for pumping the trapped volume of gas. Figure 2b shows a better geometry that was also used. The thermocouple was mounted in the side of the electrode and a shield was installed to reduce the electric field at the mount and support tube to very low values.

Typical base pressures in the test stand were between 2 and 5 x 10-7 Torr depending on pumpdown times. Oxygen-free-high-conductivity (OFHC) copper, niobium, titanium, type 304 stainless steel and 6061 T6 aluminum electrodes were tested. The usual surface preparation was either mechanical polishing or micropolishing of both electrodes and some were tested in the machined-only state. Mechanical polishing means successive polishing of machined electrodes with wet aluminum-oxide papers to # 600 grit and often adding one more polishing step with one-half micron aluminum oxide suspended in water on a low-lint wipe. Micropolishing used the same initial mechanical polishing followed by finishing with either a sub-micron silicon oxide polish in a basic (pH = 9) solution or commercially available Brasso® (pH = 10) polish, a very fine brass polish in a hydrocarbon suspension. The interior of the test stand was also finished in a similar manner. With these surface preparations, very high electric fields could be reached before any measurable field emission occurred [14] and tests of gas desorption phenomena could be made independent of competing effects from field emission.



Figure 2. Two configurations of a grounded electrode that was capable of being thermally cycled to temperatures of about 600oC.

Figure 3 shows another configuration of a grounded electrode designed to produce a controlled avalanche discharge in the high-voltage gap. Gas, usually nitrogen or air, was metered through a small (1/2-mm diameter) hole in the center of the test electrode. This could be located at a controlled gap across from either a cathode or anode mounted on the high-voltage insulator. Also shown on Figure 3 was a configuration used for several experiments that permitted the electrode to be spaced about ½ cm from the thermal mount. This configuration permitted the mount and thin support tube to be heated through a thermal cycle with the electrode decoupled from the heat. This was used to demonstrate that x-rays measured during typical tests were not from the support structure.



Figure 3. Configuration of the grounded electrode used for avalanche-discharge experiments.

# 3. Review of Previous Work

A number of experiments on gas desorption were reported in a previous publication [15]. Two of the most important results are reviewed here to provide the demonstration of quantities of gas desorbed during high-voltage conditioning and the origin of the gas desorption.

**3.1 Double-Negative Experiments**

The configuration shown in Figure 1 was used with both high-voltage supplies used as negative. Numerous experiments had demonstrated desorption of gas during high-voltage conditioning but it was not possible to determine the source of desorption from those experiments [15]. In the double-negative experiments, both negative supplies were slowly conditioned to -100 kV with the voltage between two polished but unconditioned electrodes at near zero. After the final voltage was reached, the two supplies were left at that voltage for a period of about 30 minutes until all conditioning activity such as x-rays and desorption were quiescent. One supply was then turned down in increments of 5 kV while x-ray activity and the system pressure were monitored. Figure 4 shows the results of the experiment [15] using 25 mm diameter titanium electrodes at a 4-mm gap. The x-axis is the time as recorded using a chart recorder as the voltage is lowered on one supply and the y-axis is the system pressure as the voltage is being changed. The actual voltage difference is shown near the top of the graph. Once the obvious conditioning activity settled, there was no measureable x-ray or vacuum activity at 95 kV over 4 mm. The square near the upper right corner is an area equivalent to one mTorr-l of gas. This is calculated using the area of pressure above the base pressure times time (Torr-s) times the measured conductance of 70 l/s. The total gas desorbed during this experiment was about 9 mTorr-l or nearly 3 x 1017 molecules from an area of between 5 cm2 if the desorption were from only one front surface, up to about four times that if the desorption were from both front surfaces and the sides of each electrode. One monolayer is usually given as about 1 x 1015 molecules/cm2 for light molecules so this quantity of desorbed gas represents a large number of monolayers.

 

Figure 4. Test stand pressure vs. time as the voltage is increased between two titanium electrodes at a 4-mm gap. The actual voltage between the electrodes is noted near the top of the figure.

**3.2 Test of Desorption Between One Conditioned and One Unconditioned Electrode**

An experiment was designed to try to understand which electrode initiated desorption of gas during high-voltage conditioning [15]. Figure 5 shows the experimental setup used. Micropolished copper electrodes were mounted on both high-voltage electrodes with the polarities shown. A third electrode was mounted on a movable, insulated tube on the side of the test stand that was grounded through 1 kΩ. This electrode was made from oxygen-free copper with the same micropolished surface preparation used for the two in-line electrodes. The side-mounted electrode was moved to the position shown in Figure 5 while the two in-line electrodes were conditioned to high voltage at a gap of about 3 mm. Each electrode and insulator was conditioned to 100 kV and operated there for 20 minutes with the other electrode at zero volts. No field emission was detected during this process. The positive electrode was then moved back to a predetermined position such that a gap of 2.7 mm (measured after the conclusion of the experiment to avoid electrode contact before data was taken) would exist between it and the side-mounted electrode. The positive high-voltage electrode was reconditioned to 100 kV in this location before the side-mounted electrode was moved into position between the two conditioned electrodes. These procedures were designed to ensure that gas desorbed during the next phase of the experiment would come only from the unconditioned surfaces of the side-mounted electrode. High-voltage was applied with one power supply while the other was left at zero volts. Power supply current and test stand pressure were monitored during this process.



Figure 5. Experimental set-up to measure desorption between a conditioned electrode opposite an unconditioned electrode.

Figure 6 shows the results of the changes in pressure as the voltage was increased in two different experiments. The data were recorded on a strip chart recorder and the x-axis is a measure of time. The numbers above the x-axis indicate the voltage used at a given time. The y-axis shows the pressure in the test stand. The dashed line shows the results for an unconditioned cathode and a conditioned anode. Clearly, there was no change in pressure throughout the entire process until the voltage reaches 95 kV at which point some desorption occurs. A field of 35 MV/m was reached in less than 200 seconds with an unconditioned cathode and with no indication of gas desorption, field emission or any other conditioning activity – a remarkable result.



Figure 6. Conditioning activity as measured by system pressure as the voltage was increased in two different experiments with one conditioned electrode and one unconditioned electrode.

The solid line shows the data when the cathode was previously conditioned and the anode was unconditioned. This data is similar to that shown in Figure 4. Large quantities of gas were desorbed at each increment starting from about 55 kV over a 3.5 mm gap. The voltage was decreased slightly at 175 seconds and the conditioning activity stopped until the voltage was turned up to the previous level of conditioning. This figure demonstrates one of the most important observations about gas desorption during high-voltage conditioning. **The gas is desorbed from the anode!**

This experiment also demonstrates that the product of the secondary emission coefficients "A" x "B" being greater than unity is not a satisfactory explanation of microdischarge activity because conditioning one electrode should alter one of the coefficients. The conditioned cathode had no measurable field emission at 100 kV over 3 mm, with a detection sensitivity of about 20 pA. Since field-emitted current increases rapidly with field, any possible undetected field emission would be below the picoampere level at the first stages of gas desorption. It is therefore unlikely that field emission was the precursor to gas desorption.

## 4. Desorption During High Voltage Conditioning

**4.1 Desorption During High-Voltage Conditioning on the Test Stand**

Figures 7 and 8 shows the results of increasing the voltage between two 36-mm diameter micropolished copper electrodes at a gap of 5 mm [15]. The x-axis shows time and the y-axis shows the partial pressure of masses 2, 15 and 32 (Figure 7) and masses 18, 28 and 44 (Figure 8). The voltage was increased from about 60 kV to 75 kV during the time from 0.3 to 1.2 minutes while attempting to maintain a reasonably steady pressure increase during that period. The fluctuations represent unavoidable pressure changes during the sampling. Since the shape of the pressure-time graphs of each desorption product is nearly rectangular for most of the time, the heights of the peaks provide a reasonable estimate of the fraction of the gas desorbed by each of the measured gases. The six peaks have been summed and the last row shows the fraction of each peak of the total desorbed gas. The six peaks listed should account for greater than 80% of the total desorption with other light hydrocarbons accounting for most of the remaining desorption.

Table 1. Major sources of desorption during high-voltage conditioning of micropolished copper electrodes.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Peak | 2 | 15 | 18 | 28 | 32 | 44 |
| ID | H2 | CH3 | H2O | CO + C2H4 | O2 | CO2 + C3H8 |
| AmplitudeX 10-7 Torr | 1.3 | 1.0 | 3.0 | 8.0 | 0.2 | 3.1 |
| % of Total | 8 | 6 | 18 | 48 | 1 | 19 |

**4.2 Desorption During High-Voltage Conditioning of a Cyclotron Deflector**

The RGA was also used to monitor the conditioning process of the electrostatic deflector [18] of the TASCC superconducting cyclotron. The deflector used a 35 cm long x 2.5 cm high negative high-voltage electrode at a gap of 5 mm from a stainless steel septum and 1 to 2 cm from other grounded surfaces that were also made from stainless steel. It was conditioned, in 5 kV increments, from about 40 to 80 kV after it was re-polished and rebuilt. Each increase in voltage beyond 40 kV produced large quantities of desorbed gas for about one minute and conditioning activity continued for many minutes after the initial desorption. Figure 9 shows the output of the RGA for one of those measurements. The traces for mass 28 and 44 are listed as CO and CO2 but each may also have a large contribution from a light hydrocarbon. As in Figures 7 and 8, the largest increase over background occurred in mass 28 (mostly an increase in CO and light hydrocarbons, it is unlikely that nitrogen contributed much of the increase) and mass 44 (mostly CO2 or a light hydrocarbon). There is a reasonable fractional increase in both molecular and atomic hydrogen, but the intensity is not as large as in the experimental results shown in Figures 7 and 8. It is not possible to make any quantitative analysis of this data because of the complex path from the centre of the cyclotron to the location of the RGA and there are two different vacuum pumps in different locations. Table 2 shows the areas under each peak taken from a linear output from the RGA as the voltage was increased from 48 to 53 kV. The area is given in units of 10-7 Torr-s. It is not possible to convert that to a quantity of gas because of the unknown pumping speed. Large quantities of water vapour and mass 28 were desorbed compared to the tests in the test stand using copper electrodes. Note however that the anode was stainless steel rather than copper.

Table 2. Desorption products during high-voltage conditioning of the electrostatic deflector of the TASCC cyclotron. The voltage was increased from 48 to 53 kV for this measurement.

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Peak | 1 | 2 | 12 | 18 | 28 | 32 | 44 | 55 |
| ID | H | H2 | C | H2O | CO + C2H4 | O2 | CO2 + C3H8 | Hydro-carbon |
| ∫ P x tX 10-7 torr-s | 0.85 | 4.6 | 7.5 | 345 | 405 | 38 | 60 | 12 |
| % of Total | 0.1 | 0.5 | 0.9 | 39.5 | 46.4 | 4.4 | 6.8 | 1.3 |

 A similar spectrum of desorbed gases was observed each time the voltage was increased and there was no obvious activity after a few minutes of conditioning. The RGA traces showed no activity during overnight conditioning at a fixed voltage and then an immediate increase similar to that shown in Figure 9 once the voltage was increased again.

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Figure 7. The partial pressures of mass 2, 15 and 32 during high-voltage conditioning of two micropolished copper electrodes.



Figure 8. The partial pressures of mass 18 (water vapour), 28 and 44 during high-voltage conditioning of two micropolished copper electrodes.



Figure 9. RGA spectrum of gases desorbed as the voltage of the electrostatic deflector was increased from 70 to 75 kV during the initial conditioning.

# 5. Thermal Desorption Experiments

**5.1 Thermal Desorption of Electrodes and the Holder**

A number of experiments were done to measure the quantity and spectral composition of gas desorbed from the same electrodes used for the high-voltage conditioning experiments. The configuration shown in Figure 2a was used for a number of those measurements and the temperature recorded was the holder temperature rather than the actual electrode temperature. Electrodes with high thermal conductivity such as copper will track much closer to the holder temperature than those with low thermal conductivity such as titanium and niobium. The electrode holder can contribute some background to any measurement with an electrode so several tests were done with the holder to determine what contribution it might produce. Figures 10 and 11 show thermal desorption spectra from the first thermal cycle of the new electrode holder with no electrode attached (Figure 10) and during a second thermal cycle without venting the test stand. Most of the gas desorbed from the stainless-steel holder is water vapour with moderate yields of masses 28 and 44. Table 3 shows the integral under each of the curves plus mass 39, representative of light hydrocarbons. The integral for all of the measurements of desorption peaks reported in the tables in this paper is the area above a base line drawn from the initial value of the pressure (30 x 10-8 Torr for the water vapour in the example shown in Figure 10), to the value of the pressure at the end of the measurement period. Water vapour is the dominant peak followed by hydrogen and masses 28 and 44. Mass 39 was present at just over one percent.



Figure 10. Thermal desorption spectrum from the first thermal cycle with the target holder to a temperature of about 550oC.

During the second thermal cycle there was very little desorbed except hydrogen that was likely desorbed from the bulk. The integral of pressure times time for hydrogen is about 50% compared to hydrogen desorption in the first thermal cycle. After a few more thermal cycles there was very little hydrogen detected from the holder.

Table 3. Integral of gas desorbed from the new stainless-steel electrode holder during a thermal cycle to about 550oC.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Peak | 2 | 18 | 28 | 39 | 44 | Total |
| ID | H2 | H2O | CO + C2H4 | C3H3 | CO2 + C3H8 |  |
| ∫ P x tX 10-7 torr-s | 567 | 1750 | 468 | 45 | 513 | 3343 |
| % of Total | 17 | 52 | 14 | 1.3 | 15 |  |



Figure 11. Thermal desorption spectrum from the second thermal cycle of the electrode holder with no venting of the test stand, to a temperature of about 550oC.

Figure 12 shows the results of tests done on the electrode holder to re-measure (see reference 15 for the first test) desorption from the electrode holder with several different treatments typical of preparation of electrodes for use as high-voltage electrodes. The plus signs and solid black line show desorption from the holder during a re-heat after sitting in the evacuated chamber for several days. The x-axis is time after turning on the heater at t = 0 and the measured temperature is shown under the x-axis. The y-axis shows the pressure increase measured at ion gauge # 2. This background can be from gas absorbed from the vacuum chamber back into the holder and also from heating surfaces of the chamber with a strong internal heat source.

The red square symbols were produced when the holder was removed from the vacuum chamber while the chamber was purged with helium and five cm of the holder was submerged in distilled water, blown dry and re-installed and pumped. The data shown is the measured data minus the background curve. Nearly all of the gas desorbed was water vapour, as expected, and the total amount desorbed was equivalent to several monolayers if there were uniform desorption from the five cm of the holder and support tube. Because there was a temperature distribution along the tube from the value shown on the x-axis of the figure that represents the holder temperature measured by the thermocouple all the way to room temperature near the flange that supports the holder, it is hard to assign an accurate desorption rate. Similar results (not shown) were found when the experiment was repeated with ethanol but the main desorption product was still water.

The (orange) circles show the same experiment when five cm of the holder was polished with wetted #600 aluminum-oxide paper, pumped and re-tested. The total quantity desorbed was more than 5 times the amount desorbed from just wetting the surface with water. Similar results were obtained from a newly machined surface without the polishing and even more was desorbed when the holder was micropolished. This result is highlighted because most metals used for high-voltage electrodes are polished in a manner similar to this before use.

The residual gas analyzer was used to record the evolution of the main peaks of gas desorbing as the tube and holder was heated. The RGA data is not shown but is summarized in Tables 3, 4 and 5 that show the integrals of the main peaks over the 12 minutes of the thermal cycle. The quantity shown in the second line of each table is the integral of the pressure increase x time (x 10-7 torr-s). The RGA was not calibrated so it cannot be used to give a measurement of the total throughput of each gas. Table 4 shows the results for the test with the holder dipped in water, blown dry and re-pumped. Most of the desorbed gas is water vapour as expected while carbon monoxide accounts for 13%.

Table 4. Integral of gas desorbed from the stainless steel electrode holder during a thermal cycle to 500oC. The holder was dipped into water, blown dry and re-pumped.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Peak | 2 | 18 | 28 | 29 | 44 | Total |
| ID | H2 | H2O | CO + C2H4 | C2H5ethanol | CO2 + C3H8 |  |
| ∫ P x tX 10-7 torr-s | - | 365 | 58 | 11 | 5.6 | 440 |
| % of Total |  | 83 | 13 | 2.5 | 1.2 |  |

Table 5 shows the same information when the five cm of the holder were polished with #600 aluminum-oxide paper, cleaned and re-pumped. The integral of water desorbed was about twice as great as in the previous experiment but there is much greater desorption of CO, CO2 and ethanol (used as the last cleaning solution). There was very little hydrogen desorbed because the holder had been through many thermal cycles by this time and most of the bulk hydrogen was likely depleted.

Table 5. Integral of gas desorbed from the stainless steel electrode holder during a thermal cycle to 500oC. Five cm were polished with #600 aluminum-oxide paper, cleaned and re-pumped.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Peak | 2 | 18 | 28 | 29 | 44 | Total |
| ID | H2 | H2O | CO + C2H4 | C2H5 orethanol | CO2 + C3H8 | Total |
| ∫ P x tX 10-7 torr-s | - | 700 | 175 | 170 | 292 | 1337 |
| % of Total |  | 52 | 13 | 13 | 22 |  |

Table 6 shows the same information when five cm of the holder were polished with #600 aluminum-oxide paper, micropolished with Brasso, cleaned and re-pumped. There is significantly more desorbed gas of each of desorption products and mass 28 shows a larger fraction of desorption.

Table 6. Integral of gas desorbed from the stainless steel electrode holder during a thermal cycle to 500oC. Five cm were polished with #600 aluminum-oxide paper, micropolished with Brasso, cleaned and re-pumped.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Peak | 2 | 18 | 28 | 29 | 44 | Total |
| ID | H2 | H2O | CO + C2H4 | C2H5 orethanol | CO2 + C3H8 |  |
| ∫ P x tX 10-7 torr-s | 23 | 1215 | 450 | 58 | 325 | 2071 |
| % of Total | 1.1 | 59 | 22 | 3 | 16 |  |



Figure 12. The gas desorbed from the stainless-steel holder as it was heated through a thermal cycle to 500oC and cooled to near room temperature.

**5.2 Thermal Desorption Opposite a Cathode at 40 to 50 kV**

Many measurements were made with the electrode undergoing thermal desorption used as an anode while a polished cathode was used on an insulator at gaps of two to six mm with a voltage of 35 to 50 kV, and a Geiger counter was used to monitor x-ray activity at the face of the glass window. One of these measurements was reported in reference 15 and Figure 13 shows that data. The +-signs show the radiation produced as a niobium electrode polished with wet #600 aluminum-oxide paper was heated during the first thermal cycle. This experiment was done using the configuration of Figure 2b with the shield in place and the thermocouple mounted in the side of the electrode. There was a broad peak in x-ray activity over a temperature range from 150 to 350oC and then as the temperature increased above about 450oC, the radiation increased exponentially with temperature. When the same electrode was re-heated without venting the test stand, the data shown as (blue) diamonds was measured. The x-ray activity remained at background until temperatures in excess 450oC and then the same exponential increase in x-ray activity as measured during the first thermal cycle was observed. These observations of radiation during a thermal cycle were unexpected and it was suggested [15] that the radiation produced at the lower temperature was produced from desorption of bursts of gas that led to an avalanche discharge and source of the other radiation remained unknown.

These interesting results led to many other experiments to try to understand this observation. A number of tests were done on the stainless steel electrode holder without any electrodes and substantial radiation was observed at temperatures above about 550oC. There was concern that some of the tests with different metals on the stainless steel holder were measuring results for the holder and not the electrode. The configuration shown in Figure 2b was developed and used for a number of tests. The outer shield reduces the electric field at the stainless steel holder to very low values so it should not contribute to the production of radiation during a thermal cycle.



Figure 13. The x-ray yield between a polished niobium anode electrode as it was heated through a thermal cycle to above 500oC and a cathode mounted on an insulator at a distance of 3 mm.

Figure 14 shows the results of thermal cycles with the two different configurations that demonstrates the effectiveness of the new set-up. In the first experiment the stainless-steel holder with no electrode was heated using the configuration shown in Figure 2a. In the second experiment the shield was installed as shown in Figure 2b and a polished electrode was mounted on a longer threaded rod (see insert on Figure 3) so that there was poor thermal contact with the holder. The x-axis shows the temperature of the holder and the y-axis shows the radiation yield measured with the Geiger counter. The +-signs show the data from the stainless-steel sample holder with no electrode, measured using the configuration shown in Figure 2a with a cathode at 3 mm and 35 kV. The exponential increase begins at about 550oC, higher than observed in the experiment shown in Figure 13. The diamond-symbols and dashed line show the results when the holder was heated using the shield and an electrode mounted with poor thermal contact. There was no increase in radiation as the holder was heated to about 650oC, giving some confidence in results such as shown in Figure 13.



Figure 14. Measurement of radiation from the stainless-steel holder and an electrode thermally isolated from the holder.

Figure 15 shows another x-ray experiment with micropolished copper electrodes using the configuration shown in Figure 2b. The solid line and (black) plus symbols show the results of heating a copper electrode with a cathode at 3 mm and at 45 kV. This was the first thermal cycle and the results are similar to that shown in Figure 13 but the transition to an exponential increase in radiation occurred at a higher temperature. The copper electrode was removed, re-polished and used in the same configuration with a 90V battery in series with a picoampere meter. There was no measured current change until about 450oC and then the current increased with the same exponential slope as the radiation graph.



Figure 15. The x-ray yield between a micropolished copper anode as it was heated through a thermal cycle to above 500oC and a cathode mounted on an insulator at a distance of 3 mm and a second thermal cycle using a re-polished anode and a cathode at 90 volts to measure current.

**5.3 Correlation of Hydrogen Desorption and Radiation**

An increase in x-rays as shown in Figures 13 and 15 was observed in many other experiments with copper, niobium, titanium and stainless-steel electrodes. The increase in x-rays occurred at about the same temperatures as hydrogen desorption began to increase. Experiments were done to try to check for correlations of the two. Figure 16 shows one of those experiments. In this experiment, a micropolished niobium electrode was used instead of the mechanical-polished electrode used for the data of Figure 13, using the configuration of Figure 2b. The left-hand y-axis shows the radiation measured with a Geiger counter at the glass window while the right-hand y-axis shows the partial pressure of H2 measured with the RGA. There is a strong correlation of the exponential increase in radiation and desorption of hydrogen. There is also the other interesting feature that there is significant radiation production in the second thermal cycle at temperatures below the exponential increase. However, there is also significant hydrogen desorption at these lower temperatures during both thermal cycles. The micropolishing with a very fine abrasive suspended in a hydrocarbon fluid tended to add more hydrogen to the surface of all metals tested and that was the likely reason for the higher amounts of hydrogen desorbed compared to the first test reported in Figure 13. However, these interesting results added significant uncertainty to the postulate that the radiation at lower temperatures was from an avalanche discharge [15]. Section 5.4 shows another very interesting observation from this experiment.



Figure 16. Possible correlation of radiation production with hydrogen desorption for a micropolished niobium electrode.

A third thermal cycle was done with this configuration, again without venting the test stand. Figure 17 shows the results of that experiment. The results of the third thermal cycle are nearly identical to the second thermal cycle.

With reference to Figure 15, several things can be estimated based on that data. The radiation produced in the temperature range from 200 to 400oC is high enough that it should have been possible to measure current at 90 volts if the radiation and current produced during the exponential region are used as a guide. It would appear that the radiation is produced under the influence of the strong electric field and not current that can be measured with the low-voltage set-up. Above about 450oC, the slope of both current (at 90 volts) and radiation are essentially the same. A very crude calculation can be done by assuming a pumping speed for hydrogen of 70 l/s and using a partial pressure of hydrogen near peak current as one to two x 10-7 Torr for a throughput of about 1 x 10-5 Torr-l/s. This is equivalent to about 3 x 1011 molecules per second. 5 nA = 3 x 1010 electrons or ions per second. Within the rough assumptions of this calculation the current measured at 90 volts is a significant fraction of the hydrogen desorbed.



Figure 17. Possible correlation of radiation production with hydrogen desorption for a micropolished niobium electrode comparing the second and third thermal cycle without venting the test stand.

Figure 18 and 19 show similar data for a stainless-steel electrode (Figure 18) and a micropolished copper electrode (Figure 19). The stainless steel electrode had been machined to a fine finish but no other polishing was applied. During the first thermal cycle a transition to field emission occurred and the radiation data was not usable. During the second thermal cycle the gap was increased a bit to remove field emission. There was also some radiation present at temperatures below the exponential region and this also correlates with a modest increase in hydrogen. Above the transition there is a good agreement in the slopes of the two processes.

Figure 19 shows a micropolished copper electrode during the first thermal cycle. There was the same modest increase in radiation near 300oC that was observed in the data shown in Figure 17 and then the exponential increase at about 550oC. The saved RGA spectrum was recorded with a range that did not measure the hydrogen until the approximate value shown. Again, there is a reasonable correlation with the two measurements.



Figure 18. Possible correlation of radiation production with hydrogen desorption for a machined stainless-steel electrode.



Figure 19. Possible correlation of radiation production with hydrogen desorption for a micropolished copper electrode.

**5.4 Comparisons of Desorption Spectra With and Without a Strong Electric Field**

 **5.4.1 Tests Using Copper Electrodes With No Electric Field Present**

Comparison of the RGA spectra from the two different thermal cycles used in Figure 15 showed an unexpected result that led to examination of several other similar cases. Figures 20 shows the RGA spectra from a new micropolished copper electrode that was heated to about 600oC with no electric field present. The electrode was heated using the configuration shown in Figure 2a and the temperatures shown are the holder temperature rather than the temperature of the actual copper electrode as used in Figure 18. The measured holder temperature is somewhat higher than the actual electrode temperature.



Figure 20. Thermal desorption from a micropolished copper electrode. No cathode was used during this desorption cycle.

Table 7 shows the integral of the various peaks over the total thermal cycle and the last row shows the fraction each peak represents of the total integral. The peaks shown represent about 90% of the total desorption. Spectra taken at high temperatures showed many light hydrocarbon peaks with low peak intensities at masses up to about 65 AMU. A large amount of both water vapour and hydrogen are present in the spectrum shown in Figure 20. The graph shown is similar to desorption spectra when the copper electrode was opposite a cathode at 90 volts (Figure 15) and another test with a new machined copper electrode with no polishing. All demonstrated the large desorption of hydrogen and water and the micropolished electrode desorbed greater amounts of mass 28 and 44.

Many desorption experiments were done with OFHC copper electrodes. In this sequence of experiments, the test stand was vented and purged with helium while the electrode was removed to air for 30 seconds using vinyl gloves, re-installed and pumped before a second thermal cycle. Table 8 shows some results from that experiment. The total quantity of gas desorbed is about 20% compared to the first thermal cycle with a micropolished electrode. There were still large quantities of water vapour and hydrogen and an increased fraction of mass 28.

Table 7. Desorption products (to about 600oC) from a micropolished copper electrode with no electric field.

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Peak (AMU) | 2 | 12 | 15 | 18 | 28 | 32 | 44 | 55 | Total |
| ID | H2 | C | CH3 | H2O | CO + N2 | O2 | CO2 |  |  |
| ∫ P x tX 10-7 torr-s | 2700 | 14 | 26 | 2900 | 180 | 24 | 290 | 19 | 6153 |
| % of Total | 44 | 0.2 | 0.4 | 47 | 2.9 | 0.4 | 4.7 | 0.3 |  |

Table 9 shows the same information for the electrode re-heated without venting to atmosphere. There is only a few percent of the total desorbed in the first test, mostly water vapour and a small amount of hydrogen. The water vapour may be more from desorption from the test stand with the strong internal thermal source than the electrode and holder because it occurred near the highest temperatures.

Table 8. Desorption products (to about 600oC) from a micropolished copper electrode during a second thermal cycle. The electrode was removed to air for 30 seconds between thermal cycles and re-pumped. No electric field was present.

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Peak | 2 | 12 | 15 | 18 | 28 | 32 | 44 | 55 | Total |
| ID | H2 | C | CH3 | H2O | CO or C2H4 | O2 | CO2 orC3H8 |  |  |
| ∫ P x tX 10-7 torr-s | 340 | <5 | <5 | 600 | 270 | 20 | 60 | <5 | 1300 |
| % of Total | 26 | - | - | 46 | 21 | 1.5 | 4.6 | - |  |

Table 9. Desorption products (to about 600oC) from a micropolished copper electrode with no electric field during a third thermal cycle, following the thermal cycle of Table 9, without venting the test stand between tests.

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Peak | 2 | 12 | 15 | 18 | 28 | 32 | 44 | 55 | Total |
| ID | H2 | C | CH3 | H2O | CO + N2 | O2 | CO2 |  |  |
| ∫ P x tX 10-7 torr-s | 11 | - | - | 72 | - | - | - | - | 83 |
| % of Total | 13 |  |  | 87 |  |  |  |  |  |

**5.4.2 Tests Using Copper Electrodes With an Electric Field Present**

Figure 21 shows the thermal desorption from the same copper electrode used for the experiments in Section 5.4.1, micropolished before use, with a cathode present at a 6 mm gap and operated at 45 kV with respect to the anode. The spectrum is dramatically different than that shown in Figure 20. The hydrogen has been reduced by a large amount and carbon monoxide (and C2H4) and especially carbon dioxide (and C3H8) has increased by a large amount. Table 10 shows the same analysis used for Table 8. The integral of the amount of water released remains about the same but the hydrogen peak has been reduced by more than a decade while the mass 44 peak has increased by a similar ratio.

Table 10. Desorption products (to about 600oC) from a micropolished copper electrode with cathode at 45 kV.

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Peak | 2 | 16 | 18 | 28 | 29 | 32 | 41 | 44 | Total |
| ID | H2 | CH4 orO | H2O | CO or C2H4 | CH3CH2 | O2 | C3H5 | CO2 orC3H8 |  |
| ∫ P x tX 10-7 torr-s | 100 | 100 | 1860 | 410 | 250 | 25 | 110 | 1200 | 4055 |
| % of Total | 2.5 | 2.5 | 46 | 10 | 6.2 | 0.6 | 2.7 | 30 |  |



Figure 21. Thermal desorption from a micropolished copper electrode with a cathode present at a 6 mm gap and operated at 45 kV with respect to the anode.

These results suggest that there may be complex chemistry occurring at the surface of the anode during the desorption process when a strong electric field is present. There may, for example, be catalysis on surface oxides. It appears that the large quantity of hydrogen desorbing thermally (Figure 20) has combined with surface carbon contamination to produce an array of light hydrocarbons with masses up to above 60 AMU’s.

The data showing the desorption products of copper electrodes during high-voltage conditioning (Figures 5 and 6) also show strong enhancement of light hydrocarbons and mass 28 and 44, very similar to the thermal desorption under the influence of strong electric fields. It is likely that similar complex electro-chemical behaviour also occurs during high-voltage conditioning. The peaks at mass 28 and 44 are not entirely from CO and CO2 but have a large component of the light hydrocarbons. The data in Table 7 for thermal desorption show that the sum of three peaks likely originating from light hydrocarbons (mass 12, 15 and 55) represents about 1% of the integrated desorption to about 600oC. The data in Table 10 show that the integral of three peaks (not including mass 28 and 44) that are from light hydrocarbons (mass 16, 29 and 41) is over 11% and mass 28 and 44 represents another 40% of the total desorbed gas.

The results shown in Sections 5.4.1 and 5.4.2 were repeated in a number of other experiments with similar results – there were some small differences depending on the initial treatment of the electrode surface. However, the main features shown in the experiments reported were repeated for all of the other experiments completed.

**5.4.3 Tests Using Niobium Electrodes With an Electric Field Present**

There were no other comparisons similar to the copper data shown in Sections 5.4.1 and 5.4.2 in which an electrode was used in identical thermal cycles with and without a strong electric field present. However, there were experiments in which a niobium electrode was heated through a thermal cycle with the cathode in place and operating at 40 to 50 kV. Figure 22 shows the thermal desorption spectrum from the first thermal cycle with a micropolished niobium electrode and a cathode present at a 3 mm gap and operated at 45 kV with respect to the anode. This is the thermal desorption spectrum taken during the radiation experiment reported in Figure 16. Some hydrogen begins to desorb between 150 and 200oC but the yield remains low until a large number of light hydrocarbons are desorbed at temperatures between 150 and 350oC. Once the rate of desorption of the light hydrocarbons is nearly complete then the rate of hydrogen desorption begins to increase.

Figure 23 shows the results of a second thermal cycle with the niobium electrode without venting the test stand. The hydrogen begins to desorb at about the same temperature as in the first test but rapidly increases to about five times higher than in the first thermal cycle and remains at that level until about 450oC where it begins to increase again with the same exponential slope as in the first thermal cycle. The results from the radiation experiment shown in Figure 16 indicate a higher radiation field at temperatures below about 450oC for the second thermal cycle compared to the first cycle, corresponding with the higher amounts of desorbing hydrogen.

When the copper electrode used for the experiment shown in Figure 21 was heated through a second thermal cycle without venting the test chamber, there was no significant desorption of anything below about 450oC and no radiation produced. Above 450oC, hydrogen was desorbed with the same exponential slope as shown in Figure 21 and the x-ray yield tracked the slope of the hydrogen yield.

Hakovirta [18] reported thermal desorption spectroscopy (TDS) results of high-quality niobium samples that were made of the same material used for superconducting rf cavities. He used an ultra-high vacuum system for TDS measurements that produced a desorption spectrum that contained water, CO and CO2, identified as surface peaks, desorbed in the temperature range from about 200oC to 350oC. Hydrogen also started to desorb at about 200oC and increased to a nearly constant value between 250oC and 350oC and this was identified as a surface peak. The hydrogen desorption increased exponentially at higher temperatures, peaking above 500oC. This peak was identified as a diffusion peak. The shape of the hydrogen desorption spectrum measured by Hakovirta was qualitatively similar to the hydrogen peak in Figure 23, the second thermal cycle without venting.



Figure 22. Thermal desorption during a first thermal cycle from a micropolished niobium electrode with a cathode present at a 3 mm gap and operated at 45 kV with respect to the anode.

This behaviour is consistent with the idea that the hydrogen is combining with surface carbon contamination during the first cycle and during the second cycle there is much less carbon available so the hydrogen desorption rate is higher.



Figure 23. Thermal desorption from a micropolished niobium electrode with a cathode present at a 3 mm gap and operated at 45 kV with respect to the anode during a second thermal cycle without venting to atmosphere.

**5.4.4 Other Tests**

There was no direct comparison between desorption from stainless-steel or titanium electrodes with and without a strong electric field. Tests were done with a large-diameter (38 mm) stainless-steel electrode using the configuration of Figure 2b. The larger size and mass of the electrode combined with the low thermal conductivity required higher power and longer times to reach temperatures in excess of 500oC compared to the smaller (25-mm diameter) electrodes used in other tests and the shield over the holder reached higher temperatures than in previous tests. This lead to significant increases in water and masses 28 and 44 AMU for temperatures above about 450oC that are not likely from the electrode, for both the first and second thermal cycle. The RGA spectra taken during the tests used for the data of Figure 18 showed qualitative agreement with the data for niobium up to about 450oC. The integral of hydrogen production up to 450oC (below the exponential increase in desorption) during the first thermal cycle was 65 x 10-7 Torr-s compared to over 200 x 10-7 Torr-s for the second thermal cycle without venting. Tests with titanium were also interesting with very low production of radiation. However, the titanium electrodes had been used in at least three previous tests with both high-voltage conditioning and thermal desorption in tests without the RGA. The results measured with the RGA may not be very representative of tests on new material.

# 6. Discussion

* 1. **Field Emission**

High-voltage conditioning of metal electrodes in a vacuum is a challenging technical problem. No theory has adequately accounted for all of the observed phenomena, in particular, desorption of large quantities of gas during the initial conditioning and the requirement for significant re-conditioning if the vacuum system is vented to atmosphere. The anode, cathode and supporting insulator all produce physical phenomena that can influence the maximum voltage that the integrated system can reach. The “physics” of the cathode is the better understood of the three components and yet there still remain significant open questions. Field emission, the production of electrons from a cold metal surface by the action of a strong electric field, is the main activity at the cathode. It has been extensively investigated [19] and the usual analytic description of field emission is the Fowler-Nordheim equation [20]. For high electric fields and low temperatures, the current density, j in (A/m2), from a metal point is given by:

 

1

where t(y) and v(y) are slowly varying functions of y = 3.8 x 10-5√E/φ. t(y) is close to unity and is usually ignored while v(y) has a variation with fields from 2 - 5 x 109 V/m (appropriate to field emission) given by:

 v(y) = 0.956 - 1.062 y2

E is the electric field in V/m; φ is the work function in eV. Real systems with large area electrodes and voltages over a few hundred kV are often limited to fields of less than 10 MV/m compared to fields of a few GV/m required for field emission. When this equation is applied to such large-area electrodes the electric field in the Fowler-Nordheim equation becomes:

 E = βV/d

where V is the total voltage in volts; d is the gap spacing in meters; and β is the field enhancement factor. The field enhancement factor has been determined analytically[21] for some common geometric shapes such as cones and cylinders and is typically close to the aspect ratio of the height of the projection to the tip radius.

There are several problems in using field emission from an isolated projection as the main precursor to breakdown between large-area electrodes. Measurements of broad-area cathode surfaces with scanning electron microscopes rarely identify sharp points with an aspect ratio as large as 20 to 100 that would be required to produce field emission at the macroscopic field in use. Other measurements with a scanning anode at a few kV generally only identify field-emitting sites at fields of greater than 100 MV/m. A second concern is that field emission is not present at low electric fields. As the field is increased there is an abrupt transition from no field emission to field-emitted currents of μA’s. However, there are no discontinuities in the Fowler-Nordheim equation over any range of electric field. As the electric field is increased, some physical process must occur to produce the transition from no field emission to the large observed currents. There have been attempts to associate enhanced field emission with dielectric layers on the metal surface [23]but the source of the “switch-on’ of field emission is still not well understood. A previous paper by the author addressed some of those issues [14].

Considerable improvement in surface preparation and the use of clean-rooms for assembly of the vacuum chamber and the electrodes has reduced the usual sources of field emission. Much of this work has been developed for superconducting rf cavities but can be readily transferred to dc high-voltage systems. In spite of the improvements in cathode surfaces leading to reduction in field emission in superconducting rf cavities, there are still large challenges in reaching high dc voltages such as300 to 500 kV at comparatively low fields of less than 10 MV/m.

* 1. **Insulators**

The choice of insulator and, especially, its termination at the cathode (the triple point) has important operational consequences. There does not appear to be a systematic study of the potential of gas desorption from the walls of an insulator subject to high electric fields. The use of a long insulator surface subject to potential charging from field-emitted electrons or scattered charged particles in applications such as electron guns or accelerator columns, can lead to punch through and catastrophic damage to the insulator. Segmented insulators [7] with a series of equipotential planes and internal shields that shield the ceramic from line-of-sight reception of field-emitted electrons helps to reduce punch-through of the insulator and regions of uneven charge buildup on the surface. The insulator design can have many impacts on the ultimate system performance and needs to be carefully considered in any design of a vacuum-high-voltage device. The approach used in reference 7 is probably about the optimal technique for high dc voltages, given the present understanding of the physical processes.

* 1. **Anode Conditioning**

Both the cathode and the insulator present significant challenges to the designer but the “physics” of both are reasonably understood. However, the “physics” of the conditioning anode remains the most complex aspect of vacuum-high-voltage insulation. Experiments reported in a previous paper [15] demonstrated that large quantities of gas were desorbed from an unconditioned anode as the voltage was increased between the anode and cathode but that paper did not present a sound physical basis to explain the observations. This paper attempts to provide some additional physical insight into the process although many unknowns remain. Two key observations have been made. The first is the large differences between thermal desorption (up to about 600oC) of a metal electrode and thermal desorption of that same electrode with a strong electric field present. The RGA spectral information of experiments with a strong electric field show large quantities of light hydrocarbons present that were not measured during the same experiment without the electric field and the simultaneous large reduction in the amount of hydrogen gas desorbed. The high yield of light hydrocarbons is also a feature of the gas desorbed from an anode during high-voltage conditioning. It appears that the hydrogen is combining with carbon contamination of the electrode surface to produce the hydrocarbons. It appears that increasing the temperature of an anode with a high electric field present produces similar processes as occurs if the voltage is increased without the heat. This may be a useful conditioning technique.

The other interesting new observation is that some of the hydrogen that is desorbed during the thermal desorption with a strong electric field present, appears to be charged. The hydrogen desorbed at higher temperatures appears to have an ionic component even without a strong electric field present. The experiments could not test if this also occurs during conditioning with high voltage. Experiments are proposed in which the cathode is grounded with a slit or circular aperture in the cathode and an energy or mass analyzer used behind it to measure properties of possible charged desorption products. It is not known from these experiments whether the charged hydrogen is atomic or molecular. There is a strong correlation of the radiation with the amount of molecular hydrogen desorbed (atomic hydrogen wasn’t measured in most tests) but is more probable that it is atomic hydrogen that is charged. This provides the basis of at least some of the radiation observed during the conditioning process as the ions are accelerated to the cathode and produce secondary electrons that are accelerated to the anode and produce x-rays. It could also be part of a feedback process in which secondary ions from the cathode are accelerated back to the anode as part of a regenerative build-up.

If the electrodes (at least for copper) are exposed to atmosphere, significant amounts of water vapour are absorbed and it would appear that some of the water breaks up on the surface to provide hydrogen that is desorbed along with the water in a subsequent thermal cycle (Table 9). This is likely a source of de-conditioning that is observed when a high-voltage assembly is vented.

* 1. **Recommendations for Anode Material**

These observations suggest potential material choices for the anode of a high-voltage assembly. If the two main observations noted above are confirmed efforts to reduce carbon contamination at the surface of the electrode and also to use materials for the anode with very low hydrogen desorption should prove beneficial. In recent years, vacuum systems fabricated from titanium alloys have demonstrated exceptionally low hydrogen outgassing after a modest bakeout [18]. The material used was a nearly pure titanium alloy (KS100 by Kobe Steel) with 0.35 wt percent of both iron and oxygen. It has an outgassing rate at room temperature that is several decades lower than the outgassing rate for best stainless steel surfaces after similar processing and a modest bake of less than 200oC for 48 hours. The low desorption rate for hydrogen is explained [18] to be due to the low activation energy of 0.21 eV for hydrogen in the thin α-Ti surface layer, compared to higher activation energy of 0.71 eV for hydrogen in the bulk material. The surface hydrogen is easily removed with the low-temperature bake and not easily replaced. This material should be tested with tests similar to those used in this paper. One other interesting idea for an assembly in which the vacuum chamber walls are also an anode surface (a common situation) is to do the thermal desorption that is used with most modern vacuum systems to achieve high and ultra-high vacuum conditions, after the high voltage has already been increased into the conditioning region. The thermal desorption may be more effective in this condition.

Even when best practices are followed, long conditioning times have been reported for conditioning high-voltage assemblies, requiring several hours conditioning for each small increase in voltage [7, 8]. Figure 4 in reference 7 illustrates this. The high-voltage vacuum chamber for a 500 kV photocathode dc gun for the ERL photocathode dc gun under development in Japan was made from a titanium alloy and reached a base pressure of 2 x 10-9 Pa (1.5 x 10-11 Torr) with about 7000 l/s pumping speed [7]. The pressure increase during voltage conditioning from 250 to 350 kV over 24 hours was to roughly 5 x 10-7 Pa (4 x 10-9 Torr) leading to a total desorption of about:

7000 l/s x 4 x 10-9 Torr x 86,000 s = 2.4 Torr-l, a large amount of gas for an ultra-high vacuum system.

The surface area was roughly 200,000 cm2 so the total desorbed per square centimetre was about 1.2 x 10-5 Torr-l/cm2 or 1.2 x 10-2 mTorr-l/cm2 (about 4 x 1014 molecules/cm2 or nearly ½ of a monolayer). The areas of the vacuum chamber that are subject to the higher electric fields would produce significantly higher desorption yields than this amount.

Is field emission a credible source for the gas desorption? The maximum x-ray field at 20 cm from the chamber was limited to a few µSv/h [7]. If that were produced from field-emitted electrons that terminated on the titanium anode, then the x-ray yield would be similar to the yield given in [20] at one metre from a tungsten target, filtered through 2.5 mm of copper. The lower Z target produces a lower yield of x-rays but the vacuum chamber walls adsorb less so to a rough approximation the two effects balance. The x-ray yield is given as [20]:

Y = 3 R/min/mA at one metre or 0.03 Sv/min/mA x 60 min/h = 1.8 Sv/h/mA at one metre x (1/.2)2 or 45 Sv/h/mA at 20 cm. This is some seven decades higher than observed, indicating that if the radiation were from field emission then it would be from emission current of the order of nanoamperes from a single emitter. The total power in the electron beam would be the order of mW’s. It seems unlikely that this would lead to the continuous large amount of desorption recorded. Beyond this observation, the large amount of gas desorbed must come from a large fraction of the anode surface and field emission interacts with only a very small site.

**6.5 Relationship of dc High-Voltage Conditioning to rf Processing**

Is there any relevance of these observations to rf processing? There does not appear to be any difference in the effects of field emission at dc conditions or rf conditions [22]. The time constant of barrier penetration is much less than a picosecond so that every surface of an rf cavity will be a cathode during each rf cycle at various gradients that are easily calculated. The time constant of voltage-induced gas desorption is from many microseconds for microdischarges up to many hours, much longer than the rf cycle time. However, during initial application of rf to a new room-temperature cavity there are large quantities of gas desorbed as the rf-field or pulse width is increased. This is considered rf-conditioning of the cavity and many procedures have been developed to optimize the process but there is still no solid explanation of the physics of the process. As in dc high-voltage conditioning, field emission is often credited as a major part of the process but field emitters are pretty stable and not usually present at low fields. The experiment on dc high-voltage conditioning reported in Figure 8 showed that a polished cathode with no previous high-voltage conditioning, opposite a conditioned anode, quickly reached 35 MV/m with no measureable activity. It is unlikely that cavities prepared with modern surface processing techniques should demonstrate field emission at low fields. However, every surface in an rf cavity will also be an anode surface for some fraction of the rf cycle. Because of the long time constants for voltage-induced desorption, it may be that there is an average anode field that leads to the same physical desorption that is described in this paper.

The same desorption in a superconducting rf cavity during rf conditioning would be cryogenically pumped until the cavity is warmed. Belomestnykh et al. [23], reporting on commissioning of the superconducting rf cavities for the CESR luminosity upgrade, reported that after initial conditioning of the SRF cavity, it was warmed up and the quantity of hydrogen measured was equivalent to seven monolayers of hydrogen with significant quantities of water vapour and carbon monoxide.

Figure 25 shows a schematic of a proposal to test if rf conditioning is the same physical process as occurs during high-voltage induced desorption. A SRF cavity (which has a large aperture) with valves at both ends is connected to an instrumented pumping station at room temperature and an electrode with a shape similar to that shown is inserted under vacuum from a side vacuum chamber. The electrode could be insulated in the transition through the vacuum components as illustrated (multiple layers of heat-shrinkable Teflon have been successfully used for a similar insulator) or an actual high-voltage cable with the outer plastic layer removed should also work. The vacuum hardware could also be of a larger diameter than illustrated until the transition to the cavity. The electrode will operate as a cathode and it should be possible to reach between 100 to 200 kV with a typical 1300 MHz cavity and a profiled electrode such as shown that clears the inside diameter by about 5 mm. Tests could be done with just high-voltage processing or with high-voltage processing combined with a bakeout to temperatures around 200oC. Once the processing is complete, withdraw the cathode and seal the SRF cavity before rf testing at cryogenic conditions to see if there are significantly less rf processing required.

Two experiments were reported in reference 15 that suggest that this might improve conditioning. An experiment was done in which a large (3.8 cm diameter) micropolished copper electrodes was mounted on the electrode holder shown in Figure 2a opposite a cathode of the same diameter. Gas was desorbed by increasing high voltage up to 90 kV over a 3 mm gap. A large quantity of gas (about 17 mTorr-l), similar to the measurement with titanium electrodes shown in Figure 4, was desorbed during this conditioning. The high voltage was turned off and the electrode was heated to about 500 oC. The total quantity of gas desorbed during this half of the experiment was about 13 mTorr-l. This amount was about ½ of the amount expected from previous experiments with a 25-mm diameter micropolished copper electrode. It appears that the increasing high voltage had efficiently removed a substantial fraction of the adsorbed material on the front side of the electrode.

A second experiment demonstrated that thermal desorption of an anode also conditions it. An unconditioned, micropolished copper anode was heated with electron bombardment to about 700oC, cooled, and moved into position at 2.5 mm from a cathode mounted on the high-voltage insulator. The cathode had previously been conditioned to 100 kV over 3 mm with a different anode mounted from the side port. When the thermally desorbed anode was tested, there was essentially no desorption as the voltage was increased to 100 kV.



Figure 25. Proposed experiment to desorb gas from the inside of a SRF cavity using dc high voltage.

# 7. Summary

An examination has been made of gas desorption from unconditioned electrodes of copper, niobium, stainless steel and titanium subjected to high voltage in vacuum. It has been shown that large quantities of water vapour, hydrogen, light hydrocarbons, carbon monoxide and carbon dioxide are desorbed from an anode as it is conditioned to high voltage in vacuum. The gas is often desorbed as microdischarges—pulses of a few to hundreds of microseconds. The pulses can continue for many minutes or even hours until the conditioning at a given voltage is complete and then repeat as soon as the voltage is increased. The quantity of gas released as an anode was conditioned to a high electric field such as 100 kV over 3 to 10 mm, was equivalent to many monolayers and consisted mostly of neutral molecules with an ionic component of a few percent. The charged portion of a microdischarge can be observed on an insulated anode or cathode as electrical pulses of many microamperes amplitude and hundreds of microseconds width, i.e., about 3 x 109 electrons or ions per microdischarge. Experimental evidence is presented that suggests that the electric charge measured during the microdischarge is coming from hydrogen desorbed as an ion from the anode surface. Most of the rest of the desorbed gas is composed of light hydrocarbons and water vapour. Gas desorption and charged particles are generally present during initial conditioning of high-voltage assemblies, even in situations where the vacuum chamber is the anode and has been built as an ultra- or extremely-high vacuum system.

Other experiments were completed that compared thermal desorption cycles with a high electric field to the same thermal cycle without an electric field. Thermal desorption from an anode with a nearby (3 to 6 mm) cathode at 40 to 50 kV showed much higher desorption of light hydrocarbons compared to desorption without the electric field present. At the same time, hydrogen desorption present in a desorption cycle without an electric field was reduced by a large amount (up to a factor of ten). It appears that the hydrogen combined with surface carbon contamination under the influence of the electric field to produce the hydrocarbons. Residual gas analysis of the gas desorbed during high-voltage conditioning showed similar spectra to those produced when the same electrode was heated with a high electric field present. These observations suggest that there may be complex chemistry at the electrode surface produced by the high electric field. The thermal desorption with a high electric field may produce similar physical action on an anode surface as further high-voltage conditioning without the assistance of higher temperatures. It would be valuable to test this idea because the combination of thermal desorption and a high electric field could be a good technique to gain further understanding of the complex behaviour of high-voltage conditioning. Experiments need to be performed that combine a high-voltage test stand with surface analysis equipment to measure the surface features of an anode before and after exposure to a high electric field without exposure to atmosphere.

The author has attempted to present his interpretation of this complex behaviour but is well aware that there may be others. The data presented in this and in two companion[14, 15] papers represent samples of a large number of experiments aimed at understanding the phenomena. There were many repeats of the key experimental results and other experiments were conducted to assist in reaching the present understanding of the data. The experiments have been described in sufficient detail that they can repeated by others and it is hoped that the results presented will stimulate further work along these lines.

The author does not understand the complex physics behind the observations. However, the topic of vacuum-high-voltage insulation still remains more an art than a science after many decades of experiments. It is noted that field emission was not present (at a detectable electron current limit of about 10 to 50 pA) during any of the experiments reported in this paper – it is not contributing to the observations. The data presented is based upon an extensive experimental program with many repeats of interesting results. The vacuum system was not up to modern standards of ultra-high vacuum technology and may have influenced some of the results. However, the electrode surfaces were prepared using the same polishing and cleaning procedures that are used for many systems and the challenges presented by desorption during high-voltage conditioning are still present in modern ultra- and extremely- high vacuum systems. Based upon the interesting observations reported in this paper, new tests are required with all of the materials used in this paper to confirm the results with measurements using a modern high-quality vacuum system. Continued experimental and theoretical work is required to try to obtain a more detailed picture of the phenomena at an anode surface.

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